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Date Out EFB: 1/21/81

To: Product Manager 12 Ellenberger
TS-767

From Dr. Willa Garner
Chief, Review Section No. 1
Environmental Fate Branch

Attached please find the environmental fate review of:

Reg./File No.: 464-LLT

Chemical: Chlorpyrifos-methyl

Type Product: Insecticide

Product Name: Reldan 4E

Company Name: Dow

Submission Purpose: New pesticide
for use on turf

ZBB Code: 3(c)(5)

ACTION CODE: 100

Date in: 9/26/80

EFB # 642

Date Completed: 1/16/81

TAIS (level II)	Days
61	12

Deferrals To:

______ Ecological Effects Branch

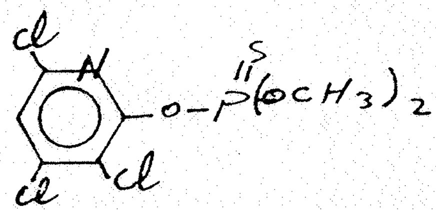
______ Residue Chemistry Branch

______ Toxicology Branch

1.0 Introduction

1.1 This submission is a 3(c)(5) request for registration of a new insecticide RELDAN 4E, containing the active ingredient chlorpyrifos-methyl. It is proposed for use as a stored grains protectant for the protection of barley, corn, oats, rice, sorghum and wheat agaist injury from stored grain weevils, moths, borers, beetles and mealworms.

1.2 Active ingredient's chemical name and structure:
0,0-dimethyl-0-(3,5,6-trichloro-2-pyridyl) phosphorothioate;



1.3 Product's commercial components: %Wt.

- Active ingredient 46.8

[redacted]

Inert ingredient information is not included

Product contains 4 lb chlorpytifos-methyl per gallon.

1.4 Active Ingredient's physical and chemical properties:

- molecular weight: 322.6
- melting point: 45.5-46.5°C;
- vapor pressure: 7.40×10^{-7} mm Hg at 0°C
  4.22×10^{-5} mm Hg at 25°C
  1.80×10^{-4} mm Hg at 35°C
- density of sp.gravity: 1.39 (50°C)
- water solubility at 23°C (g product/100 g solvent): 0.0004
  (sol. of hydrolytic metabolite 3,5,6-trichloro-2-pyridinol in water at 25°C = 0.022)
- calculated K_{ow} (Kenaga,1971): 9,300
  (for the hyrolytic metabolite, K_{ow} determined as 1620)
- physical state: granular crystalline solid
- color: white

1.5 There are only two past reviews in EFB's file for EUPs on Forest use, dated 4/15/76 and 6/22/76.

2.0 Directions for Use

2.1 Dilute RELDAN 4E with water and apply to the moving grain stream as a coarse spray to give a deposit of 3ppm to 6ppm of chlropyrifos-methyl on the grain. Final spray volume may vary from 1 to 5 gallons per 1000 bushels (0.2 to 1.0 liters per metric ton) of grain. Use the higher dosage rate if grain is to be stored for longer than 6 months.

2.2 Application rates of RELDAN 4E per 1000 bushels are:

	3 ppm		6 ppm	
	fl oz	ml	fl oz	ml
Barely	4.6	135	9.2	270
Corn	5.4	160	10.7	315
Oats	3.1	90	6.2	180
Rice	4.3	130	8.6	255
Sorghum(milo)	5.4	160	10.7	315
Wheat	5.8	170	11.5	340

2.3 If applied to metric tons, the rate of application per 100 tons is:

3 ppm		6 ppm	
fl oz	ml	fl oz	ml
20.8	625	41.7	1250

2.4 Grain Bin and Warehouse: dilute RELDAN 4E with water to give a final concentration of 1% chlorpyrifos-methyl (1/2 pint of RELDAN 4E per 3 gallons or 1/2 liter of RELDAN 4E per 25 liters). Apply spray to wall and floor surfaces at the rate of 1 gallon per 650 to 1250 ft^2 (946 ml per 60 to 115 m^2) prior to storing grain. Application should be made only after buildings have been thoroughly cleaned.

2.5 Environmental Hazards: a) RELDAN 4E is toxic to fish, birds and other wildlife; b) Keep out of lakes, streams, ponds, tidal marshes and, estuaries; c) Shrimp and crabs may be killed at application rates recommended on this label; d) Do not apply where runoff is likely to occur; e) Don not apply where weather conditions favor drift from areas treated; f) Do not contaminated water by cleaning of equipment or disposal of wastes.

3.0 Dicussion of Data

The environmental chemistry data of this submission is found in report no.7, pp.6-55 of the submitted document (EPA Acc.#099637), by Eugene E. Kenaga, July 30, 1976. The report is a compilation summarizing the data of refrenced studies (list of refrences is attached) related to the pesticide's environmental fate. Specifically, results of pesticide's metabolism and movement in soil, photodegradation in air and water and other solvents and residues in fish, mammals and in plants are discussed.

Following are synopsis of the cited results:

3.1 Hydrolysis

The major products of hydrolysis identified by Burst (1969) were 3,5,6-trichloro-2-pyridinol and phosphorothioic acids.
Hydrolysis tests were run by Meikle (1973) using low concentration (0.088 ppm w/v of 2.72×10^{-7} M) of chlorpyrifos-methyl in tap water and phosphate buffered distilled water over a range of temperatures and pH conditions:

Temp. °C	Kind of Water	pH	Half-Life Days
35	Tap	8.0	3.6
	Buffered distilled	7.8	4.8
	ditto	6.7	5.3
	ditto	4.2	7.4
25	Tap	8.0	9.4
	Buffered distilled	7.8	12.7
	ditto	6.7	17.4
	ditto	4.2	22.7
15	Tap	8.0	25.1
	Buffered distilled	7.8	45.1
	ditto	6.7	54.5
	ditto	4.2	76.0

It is noted that the presence of 0.01 ppm w/v of copper in the tap water enhanced the hydrolysis rate of chlorpyrifos-methyl at an increasing rate at 25°C as the pH decreased as follows:
pH 8 = 1x; pH 7.8 = 1.4x; pH 6.7 = 1.9x; pH 4.2 = 2.4x .
The enhanced hydrolysis rate of the chlorpyrifos-methyl is perhaps due to chelation by metals like copper which are commonly found in water.

3.2 Pesticide Fate and Movement in Soil

a) Soil Metabolism and Persistence: The aerobic degradation of chlorpyrifos-methyl [0,0-dimethyl-0-(3,5,6-trichloro-2-pyridyl-2,6-$^{14}C_2$) phosphorothioate] was studied in the lab at 1ppm in two soils (an Illinois silty clay loam containing 4.2% OC and a CA loam containing

0.8% OC) incubated at 15, 25 and 35°C and 32 or 100% moist.cap.) for up to 428 days (Regoli et al, 1974). All incubations were conducted in closed, aerated containers, and analyses of radioactivity of samples were accomplished by a combination of combustion, extraction, and TLC. The investigator reports an overall average of 100.4% recovery of added activity obtained for samples over the entire study. The major metabolite in soil was 3,5,6-trichloro-2-pyridinol, followed by lesser amounts of 2,3,5-trichloro-6-methoxypyridine (=3,5,6-trichloro-2-methoxypyridine) and $^{14}CO_2$.

It is observed in this study that the breakdown of chlorpyrifos-methyl to 3,5,6-trichloro-2-pyridinol is a rapid process if soil conditions are favorable for microbial activity (i.e. high temp. and moist. content). The time required for 50% breakdown ranged from 1.5 days in the Ill. soil at 35°C to 17.7 days in the CA soil at 15°C. Calculated times required to decrease to 10% of applied dose ranged from 29.8 to 499.7 days under the above conditions.

b) Dissipation: The dissipation of chlorpyrifos-methyl residues in various soils over a period of 30 days, after a single application of a 3% dust to Japanese rice paddy fields, was studied by Tukano and reported by Kenaga(1976a). The observed results show that the insecticide's half-life for a dosage of 1.8 Kg/ha appear to be 5-10days while a dosage of 0.18 Kg/ha had a half-life of 10-20 days.

In a lab test, when chloryprifos-methyl was applied directly to soil rather than applying it as a dust formulation) at dosage of 5ppm, it yielded a half-life of <3 days; and 0.07 and 0.3 ppm of chlropyrifos-methyl residues in soil.

3.3 Adsorption Studies as Related to Leachability

The soil adsorption of chlorpyrifos-methyl was determined by Hamaker (1974a) by analysis of both the supernatant and the soil from slurries (4 parts of 1ppm solution and 1 part soil). Measurements taken on a group of 10 soils, ranging in OC content from 0.29% to 5.76%, showed 46 to 99% adsorption respectively after 24hrs. incubation and K_{OC} of 3,304(4hrs.) and 4,585(24 hrs.). This suggests very little likelihood of significant leaching from soil treated with chlorpyrifos-methyl.

In another adsorption study Hamaker(1974b) used 3,5,6-trichloro-2-pyridinol (hydrolysis product) on three soils using an aqueous slurry method over a 3-day period that suggested a strong correlation between the degree of adsorption and soil OC where the adsorption increased with the increase in the soil's OC (18% for 0.5% OC and 68% for 3.6% OC). The reported K_{OC} value of 3,5,6-trichloro-2-pyridinol is 132.

3.4 Photodegradation

The photogradation studies of chlorpyrifos-methyl and its hydrolytic product trichloropyridinol in water and solutions revealed that they are rapidly simulated by sunlight to less chlorinated or unchlorinated derivatives, possibly dimers and/or simple basic chemical building blocks such as CO, ammonia and organic amines and acids used in plants and animals.

Smith et al(1972) exposed ring labeled ^{14}C chlorpyrifos-methyl at a conc. of 50 ppb in air at 100°F and 98-99% relative humidity to UV irradiation (300-400 nm). The data obtained indicate that the compound hydrolysis products rapidly undergo photodecomposition similar to that in water (Smith 1968). The first step in the process was the liberation of 3,5,6-trichloro-2-pyridinol, which in turn undergoes dehalogenation followed by oxidation to form a series of hydroxylated pyridinol derivatives. Further oxidation results in ring cleavage.

3.5 Distribution in Aquatic Ecosystem

The environmental distribution of chlorpyrifos-methyl and metabolites was studied by Metcalf and Lu (1973) in a model devised by Metcalf et al (1971) where sorghum was grown on a terrestrial shell of sand rising out of water in an aquarium covered to retared evaporation contianing 7L of water with mineral nutrition. The sorghum foliage was treated with ^{14}C-ring labeled compound (sp. activity 10.7 mCi/mM) at 0.2 lb/A rate and infested with larvae and other organisms and fish.
The system was exposed for a total period of 33 days to 5,000 foot candles day light equivalent at 80°F(26°C).
At the end of the exposure period, samples of fish and other organisms (algae, mosquito larvae & snails) were assayed for total ^{14}C radioactivity, extracted with acetonitrile and the extract was subjected to TLC analysis with appropriate solvents and radioautographs made of developed plates. Whenever possible standard degradation products were cochromatographed with the extracts for comparison.
Under this system conditions, chlorpyrifos-methyl was found in trace amount (8-44 ppb in the systems's organisms), 52% of the radioactivity in fish was unextractable; and 3,5,6-trichloro-2-pyridinol was found in appreciable amounts in the organisms. The biodegradability index(BI) of chlorpryrifos-methyl was calculated by dividing the concentration of the polar metabolites in substrate(organism) by the concentration of the non-polar metabolites in the substrate; BI=3.95 in this system.
In another study, 3,5,6-trichloro-2-pyridinol was applied alone to the system at 1.0 lb/A. The results of this test shows that pyridinol was stored unconjugated in all organisms, and three other degradates were found in water only.
The biomagnification factor for the two were calculated as ecological magnification(EM) by dividing the concentration of a given chemical in

The EM values for chlorpyrifos-methyl and pyridinol in fish measured 95 and 16, respectively indicating that the pesticide and its metabolite are environmentally much safer than other chlorinated hydrocarbon pesticides (EM range 287-83,500) from the stand point of buildup of residues in tissues or biodegradation.

3.6 Residues in Plants

The investigation by Leuk et al (1975) found chlorpyrifos-methyl to be relatively non-persistent in fields where coastal grass and corn were sprayed once with 0.56, 1.12 & 2.24 kg of chlorpyrifos-methyl/ha. Residues of chlorpyrifos-methyl and its hydrolysis product 3,5,6-trichloro-2-pyridinol found in grass forage 21 days after treatment ranged from <0.01 to 0.09ppm and from 0.32 to 1.24ppm, respectively; and on corn the same residues measured 0.03-0.2 and 0.09-0.6ppm. The results indicate that the hydrolysis product "pyridinol" is more persistant; while the chlorpyifos-methyl residues deminish rapidly after application.
Kenaga (1976a) reports that residue studies of many chlorpyrifos-methyl treated field crops in the US, Japan, and Near East have shown a rapid decrease in residues of the insecticide and low residues of its metabolite, 3,5,6trichloro-2-pyridinol, even after repeated treatments.

3.7 Microorganisms

MacRae et al (1974) studied the effects of ten organophosphorus insecticides including chlorpyrifos-methyl at 2 & 10ppm, on the O_2 uptake of Azotobacter vinelandii over a two hour period. At the end of the growth period, the O_2 consumption of Azotobacter v. at 2 ppm was identical to the control and at 10ppm, it meassured 71% of the control value.
Wood et al (1974) examimed the effects of chlorpyrifos-methyl and its trichloropyridinol on nitrogen fixation of Azotobacter v. employing the acetylene reduction test method. It was noted that chlorpyrifos-methyl at 500 ppm had no significant effect on ethylene; while the trichloropyridinol caused 100% reduction of ethylene production at same concentration.
Lab test using agar media containing chlorpyrifos-methyl at 500pm concentration showed no growth inhibition of the following bacterial and fungal species:
Bacillus subtilis, Pseudomonas aeruginosa, Salmonella typhosa, Staphylococcus aureus, Asperigillus terreus, Candida pelliculosa, Pullularis pullulans, and Rhizopus nigricans (Kenaga, 1976b).
Though a similar lab. test of 3,5,6-trichloro-2-pyridinol showed some inhibitory effects on the same organisms, there is no indication that microorganisms would be effected at concentrations resulting from the use of the parent insecticide.

3.8 Residues in Fish

The distribution potential of 3,5,6-trichloro-2-pyridinol-$^{14}C_2$ (pyridinol) in mosquito fish exposed for 6days in a flowing water system at 25°C was studied by Hedlund (1972) using a solution concentration of 1.1 ppb. The pyridinol concentration in the whole body of fish reached a plateau of 3.4 ppb by the third day of exposure which amounts to a very low bioconcentration factor. All radioactivity was gone from fish within 3 days after withdrawal from exposure medium indicating the likelihood of very little bioconcentration, rapid metabolism, and very quick excretion of the pyridinol in fish.

4.0 CONCLUSIONS

4.1 Hydrolysis: Chlorpyrifos-methyl is noted to have a relatively rapid rate of hydrolysis in water and soil that is enhanced by increasing of temperature, pH, and by the presence of copper irons; at pH, and by the presence of copper irons; at pH=8 t1/2 = 25.1 d (15°C), 9.4 d (25°C) and 3.6 d (35°C). It appears to hydrolyze to 3,5,6-trichloro-2-pyridinol and phosphorothioc acids.

4.2 Soil Metabolism and Persistence: In various soil, chlorpyrifos-methyl breaks down to 3,5,6-trichloro-2-pyridinol (pyridinol) with half-life varying from several days to several weeks; the process is expedite under favorable soil conditions (i.e. high temp. and moist.content) for microbial activity [t1/2 = 1.5 d at 35° and 17.7 d at 15°C]. Field testing of two dosages in two different soils yielded average residues of 0.16 & <0.05 ppm chlorpynfos-methyl for 1.8 Kg/ha and t1/2 = 5-10 d; While the residues measured 0.01 and 0.01 ppm for 0.18 Kg/ha and t1/2 = 10-20 days, on the 30th day after application.

4.3 Adsorption and Mobility: Chlorpyrifos-methyl seems to have a moderately high soil adsorption that is strong enough to result in minimal soil leaching and runoff. Pyridinol is also noted to have a moderately high partioning coefficient and soil adsorption; strong enough to prevent significant leaching.

4.4 Photodegradation: The submitted data indicate that chlorpyrifos-methyl and its major degradate, pyridinol, are degraded in water to less chlorinated derivatives and simple molecules.

4.5 Accumulation: Distribution of chlorpyrifos-methyl in Metcalf ecosystem showed only moderate accumulation in snails, mosquitoes, unspecified fish species and algae, while its hydrolysis product pyridinol did not accumulate to significent degree.
Accumulation in field crops, as reported by Kenaga, showed rapid decrease in residues of chlorpyrifos-methyl and low residues of its metabolite pyridinol even after repeated treatment.

5.0 RECOMMENDATION

5.1 In light of the above, the potential fate of chlorpyrifos-methyl in the environment has been satisfactorily identified only for the proposed use. It should be noted; however, that there are numereous data gaps in the reported information subject to questioning in conjunction with registration requests for major use patterns.

5.2 In the reviewer's opinion, EFB can concur with the 3(c)(5) registration of RELDAN 4E for use as a stored grain protectant, conditioned by the following label restriction: "Do not discharge directly or indirectly (i.e. to wastewater treatment system) to surface water". This restriction is warranted because of the potential toxicity of chlorpyrifos-methyl and its major degradate pyridinol to fish.

5.3 For any subsequent registration actions involving any of the uses identified in Table 1 of the July, 1978, proposed guidelines, applicant must submit the detailed studies of the pertinent environmental chemistry requirements in order to adequately establish the active ingredient's environmental profile and fulfill potential data gaps.

5.4 Note to PM

In the directions for use we note only one rate of application expressed in metric units, whereas several rates were identified in the other unit for the different seeds; it is not clear whether the metric rates should identify other rates.

Madeline Nawar M. Nawar 1/16/81
Environmental Fate Branch
Section #1
1/13/81

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